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The Needs for Carbon Dioxide Capture from Petroleum Industry: A Comparative Study in an Iranian Petrochemical Plant by Using Simulated Process Data

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1. Introduction

The greenhouse effect is the heating of the earth due to the presence of greenhouse gases. According to the Intergovernmental Panel on Climate Change (IPCC), the ongoing emissions of greenhouse gases from human activities are leading to an enhanced greenhouse effect. This may result, on average, in additional warming of the earth's surface (Houghton and Jenkins, 1990). During 2007, global emission of carbon was 7 billion metric tons (Bt), which are expected to increase to 14 Bt per annum by the year 2050 assuming the demand for fossil fuel keeps increasing because of the growing economies around the world (Bryant, 2007). Carbon dioxide (CO_2), is considered as a raw material in chemical industry. So, its recovery from flue gas meets a great prosperity not only for economic point of view but also for its negative effects to the environment. CO_2 emission control by its capturing from fossil-fuel combustion sources is applied widespread in power plants and industrial sectors. By utilization of this approach, fossil fuel could be continually allowed to be used with a lesser degree and/or without contributing significantly to greenhouse-gas warming.

This chapter clearly shows the need for CO_2 capture in downstream petroleum industry by demonstrating its health and environmental effects. These effects are briefly discussed the negative impacts of the increasing trend of CO_2 emission in Iran. Afterward, a comparative study for capturing carbon dioxide in a petrochemical plant in Iran will be presented.

1.1 CO_2 health effects

At 5% concentration in air (500,000 parts per million (ppm)), CO_2 can produce shortness of breath, dizziness, mental confusion, headache and possible loss of consciousness. At 10 % concentrations, the patient normally loses consciousness and will die unless it is removed. With little or no warning from taste or odour, it is possible to enter a tank or a pit full of CO_2 and be asphyxiated in a very short time. Long-term exposure at concentrations of 1-2 % can cause increased calcium deposition in body tissue, and may cause mild stress and behavioural changes. The National Institute for Occupational Safety and Health (NIOSH) air quality standard for the protection of occupational health sets the limit for CO_2 at 10,000 ppm for 10 hours. The Occupational Safety and Health Administration (OSHA) air quality standards for the protection of occupational health set the limit for CO_2 at 5,000 ppm (Webster, 1995).

Human and environmental impacts of solvent-related emissions at a capture and storage of CO_2 facility were estimated by Veltman et al. (2010). They stated that, although carbon dioxide capture is relatively well-studied in terms of power generation efficiency, CO_2 emission reduction, and cost of implementation, but little is known about the potential impacts on human health and the environment. The U.S. Environmental Protection Agency (EPA) has officially declared that CO_2 and other so-called greenhouse gases are dangerous to public health and welfare, paving the way for much stricter emissions standards (EPA, 2009). The 2009 CO_2 emission shows that the Middle East accounted for 3.3% of the total world CO_2, of which 31% is the share of Iran. Consequently, as shown in Figure 1, the trend of CO_2 emission is progressively increased from 1998 to 2009, which certainly endanger all aspects of life in Iran. The urban environment of Iran is becoming increasingly polluted, with adverse impacts on the health, welfare and productivity of the population. Results indicate that pollution in Tehran, where 20% of Iran's population lives, has well exceeded safe levels (EIA, 2000; Asgari et al., 1998; Masjedi et al., 1998).

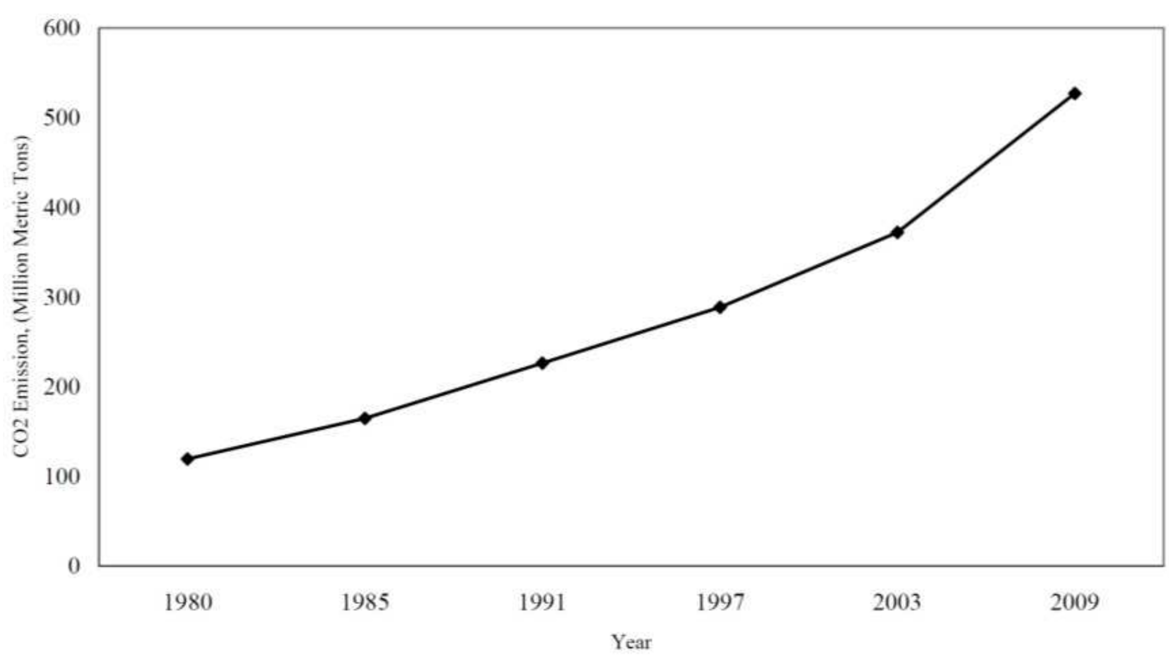

Fig. 1. Increasing trend of total CO_2 emission per Mt from 1980 to 2009 in Iran

1.2 CO_2 emission in Iran

Iran is the second-largest producer and exporter in The Organization of the Petroleum Exporting Countries (OPEC), and in 2008 was the fourth-largest exporter of crude oil globally. Iran holds the world's third-largest proven oil reserves and the world's second-largest natural gas reserves. Figure 2 shows the total fuel consumption in Iran. As it is clear in Figure 3, the combustion of fossil energy contributes with about 84 % to the CO_2 emission in Iran.

The main resource of CO_2 emission is fossil fuels that unfortunately now a day are the basic sources to generate energy in industrial-economic systems. On the other hand, energy is a main factor to achieve economic development, which is highly needed for developing countries. In 2009, Iran consists of 527 million metric tones (Mt) of CO_2 emission and is known to be the 9th worst polluter increasing the emissions by 3.2 per cent to 2009, compared with 2008 levels.

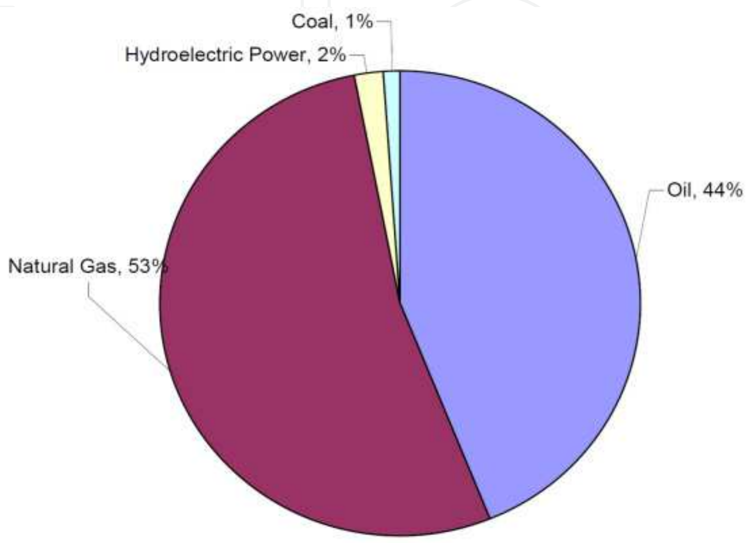

Fig. 2. Total energy consumption in Iran

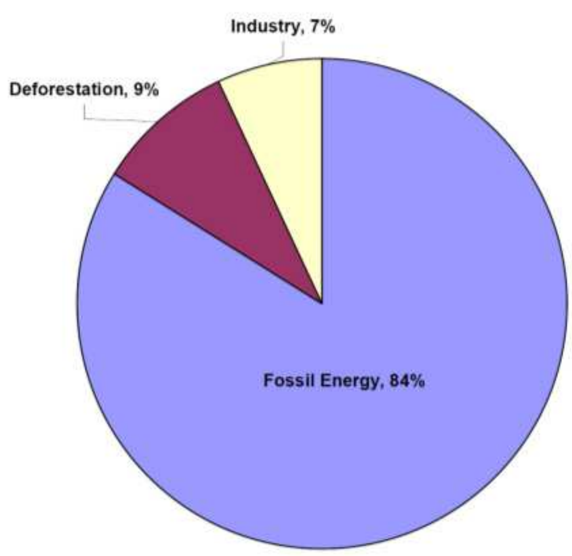

Fig. 3. Overall CO_2 emission in Iran

As shown in Figure 4, the power plant sector with an emission of 28% CO_2 is the largest carbon dioxide emissions source in Iran. Although the consumption of gas has increased in this sector during recent years, still more than 50% of the energy consumption comes from the combustion of heavy fuel oil. The industry sector is the second largest contributor to CO_2 emissions, with about 133 Mt in 2008. The transport sector accounted for 23% of total CO_2 emitted. As Figure 4 shows, the industry sector with about 26% of the total CO_2 emissions was the second major contributor in 2008. The breakdown of the industrial CO_2 emission in

Iran (Figure 5) shows that the petrochemical industry in Iran has more emission contribution in contrast with the other industries such as cement, steel, and gas processing plants. Boilers, process heaters, and other process equipment are the major CO_2 emissions producers in a petrochemical plant. The data presented in Figure 2 through Figure 5 were extracted from different literature; Moradi et al. (2008), Avami and Farahmandpour, (2008), and NIOC (2011).

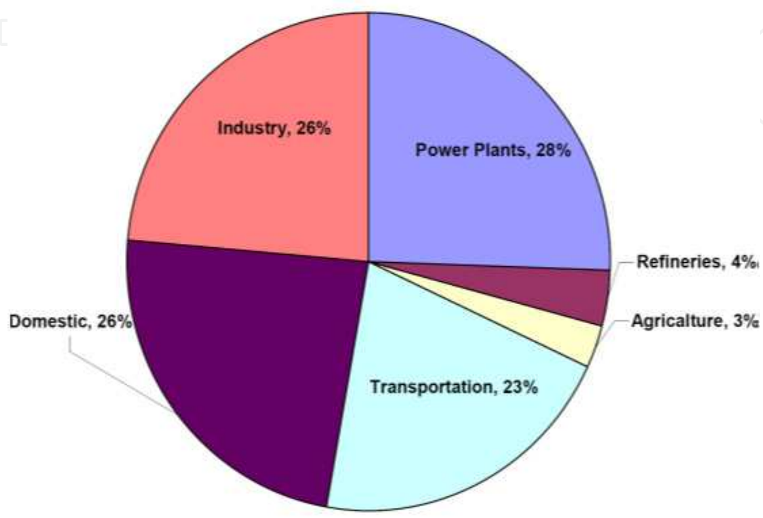

Fig. 4. Iran's Energy Sectors contributed to CO_2 emission in 2008

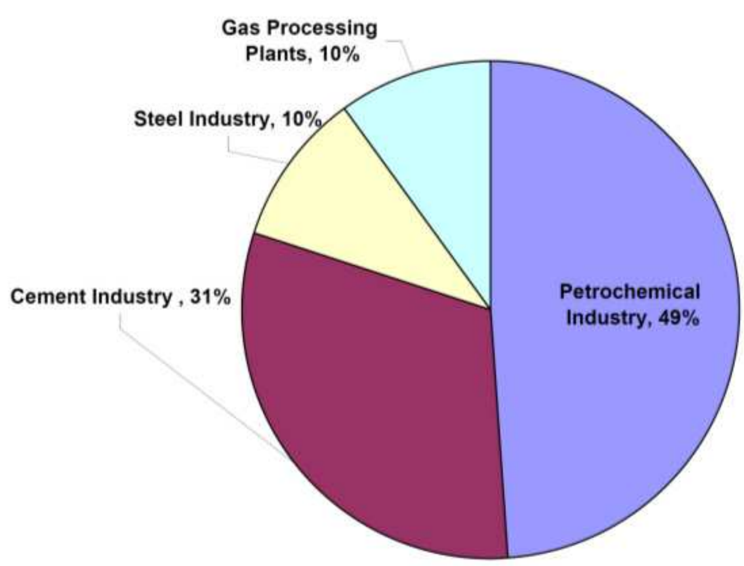

Fig. 5. The breakdown of industrial CO_2 emission in 2008

According to the recent study performed by Roshan et al. (2011), the country's temperature annual trend increment would have an increase of maximum 5.72°C to minimum 3.23°C, while considering the most optimistic case, the country's annual temperature would increased by 4.41 °C till 2100. Figure 6 shows the prediction of the total average of temperature annual and seasonal changes from 2025 to 2100 based on the results of an applied scenario for different regions of Iran. According to Figure 7, the highest amount of CO_2 density, which has been forecasted for the year 2100 is 570 ppm.

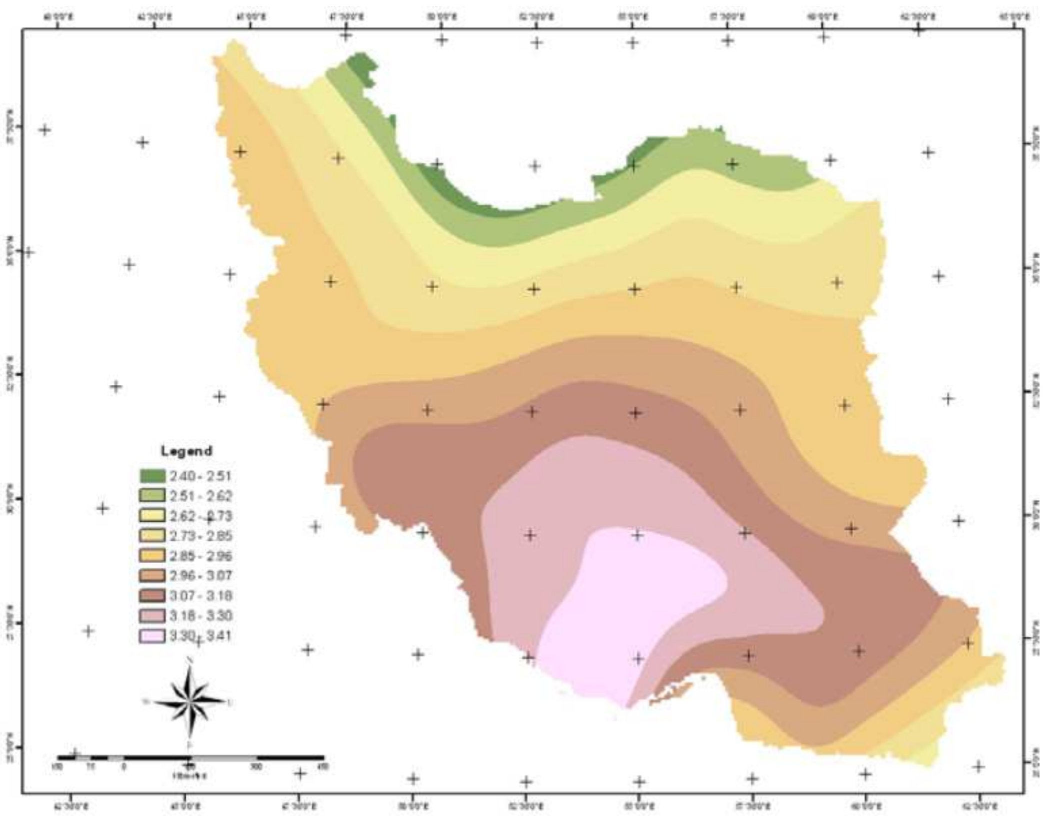

Fig. 6. Prediction of the total average of temperature annual and seasonal changes from 2025 to 2100 based on the results of an applied scenario for different regions of Iran (Roshan et al. 2011)

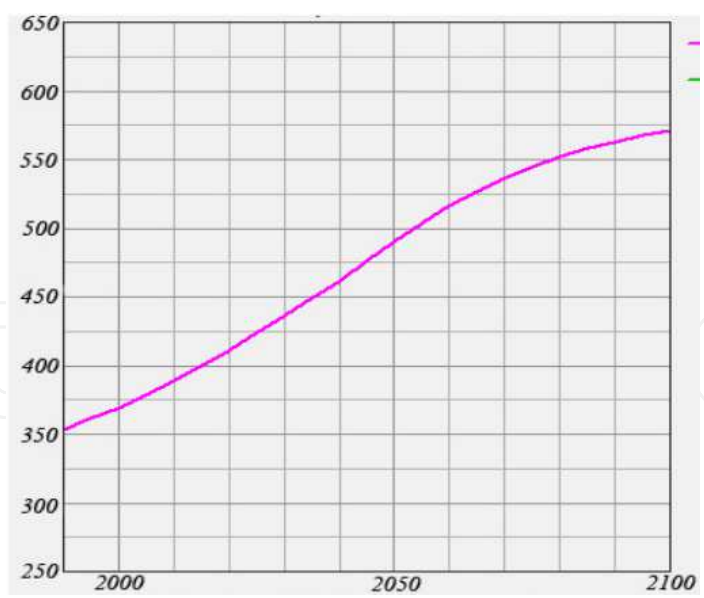

Fig. 7. Prediction of the CO_2 concentration per Mt till 2100 in Iran (Roshan et al. 2011)

2. CO_2 capture technologies

As reported by the United Nations Intergovernmental Panel on Climate Changes (UNIPCH) (1995), CO_2 level has risen 30% to nearly 360 ppm from a pre-industrial era level of 280 ppm.

Judkins et al. (1993) believed that in order to avoid major climate changes, human-generated emissions of CO_2 will have to be reduced by as much as 50-80%. As a result, three strategies had proposed for CO_2 emission control: (1) Exploiting the fuels more efficiently. (2) Replacing coal by natural gas. (3) Recovering and sequestering of CO_2 emissions.

By considering the greenhouse gas effects, it is accepted that natural gas is preferable to other fossil fuels such as coal, and oil. Indeed, removal of CO_2 from natural gas is considered as a practical and more convenience step toward reduction of CO_2 emissions. Removals of CO_2 from gaseous streams have been a current procedure in the chemical industry. Because of the increasing trend of energy consumption globally, removal of CO_2 from natural gas is not easy to be achieved; this task, obviously, required an integrated approach based on modern capturing technologies. The choice of a suitable technology (Figure 8) depends on the characteristics of the flue gas stream, which depend mainly on the chemical or power plant technology. Figure 8 shows the technologies which are currently used for CO_2 capturing.

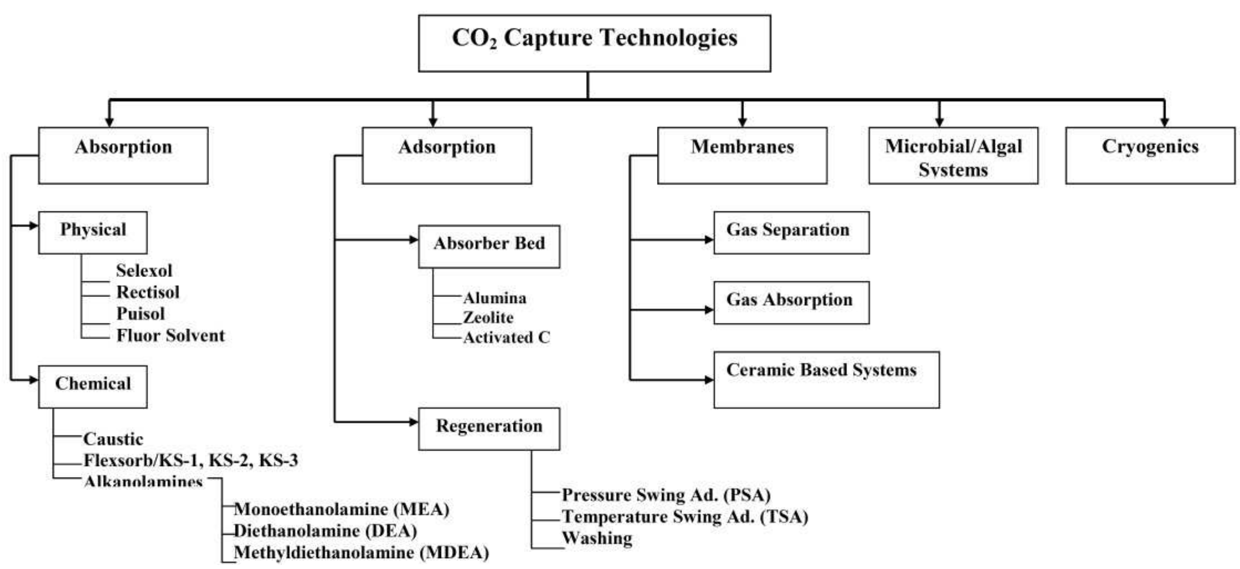

Fig. 8. CO_2 capture technologies

Chemical solvent absorption is based on reactions between CO_2 and one or more basic absorbents such as aqueous solutions of Monoethanolamine (MEA), Diethanolamine (DEA) and Methyldiethanolamine (MDEA). An advantageous characteristic of absorption is that it can be reversed by sending the CO_2-rich absorbent to a stripper where the temperature is raised.

In the present study, simulation of a CO_2 capture from fuel gases of one of the petrochemical plants in Iran was performed by using MEA, DEA and MDEA. In this work, a process by using alkanolamines including CO_2 capture from flue gases was simulated and optimized in a petrochemical plant in Iran. The simulation has been conducted using a commercial software. The required data such as the composition of three type of alcanolamines, were derived in the laboratory. This work consists of six important variables as the output of the simulation process; (1) the amount of CO_2 recovery, (2) amine consumption, (3) mechanical and operational characteristics of the absorption column, (4) CO_2 purity in the stripper column effluent, (5) required energy of the stripper, and (6) mechanical and operational features of the stripper.

In section 3, the applied process in this study for Amine-based CO_2 capture will be described. The results of this work will be presented in Section 4 and discussed in Section 5. Finally, based on different criteria, the selected alkanolamine will be demonstrated.

3. Amine-based CO_2 capture plant: Process description

Amine process is the best and commonest choice for separation of CO_2 from flue gases. Driving force of this process is the reaction between CO_2 and amine in which CO_2 with high purity is acquired by one stage process. This process starts with cooling flue gases applying a water cooler to lessen some of their impurities such as NO_x and SO_x to an acceptable value. Then the chilled gas is pressurized with a blower to the absorption column. Temperature ranges at the top and bottom of the column are about 40-45 and 50-60 °C, respectively. Flue gases and the lean amine are contacted, and CO_2 is absorbed in the amine solution through the absorber. Rich amine at the bottom of the column is pumped into a cross heat exchanger, where its temperature reaches to about 100 °C exchanging heat with the effluent fresh amine of stripping column. Then this solution introduced to the top section of the stripping column. Operating temperature at the top and bottom of the column, operating pressure and column pressure gradient are 110 °C, 120 °C, 1.3 bar and 0.17 bar, respectively.

Required energy for stripping column is supplied from saturated steam at 45 psia. The rich solution of amine and steam are contacted in stripper, and CO_2 is separated from amine. The gas stream containing CO_2 and water steam is exhausted from the top of the column to a condenser where its temperature is lowered to 45 °C. Almost the whole steam is condensed in the condenser and recycled to the top of the column. CO_2 is recovered in a flash drum, then dried and finally compressed to an acceptable pressure. The CO_2-lean solution leaves the reboiler and enters the cross heat exchanger where it is cooled. The solution is then cooled further before it re-enters the absorber.

Packed columns are often employed in the removal of impurities from gas streams and also the removal of volatile components from liquid streams. The dimensionless Robbins correlation factor is actually the Dry Bed Packing Factor issued to calculate the gas and liquid loading factors, which are in turn used to calculate the pressure drop, particularly with newer packing materials. As shown in Table 3 and Table 5, Robbins packing correlation was used as a default correlation. The Height Equivalent to a Theoretical Plate (HETP) relates to packed towers. The value refers to the height of packing that is equivalent to a theoretical plate. As shown in Table 3 and Table 5, Frank correlation was used to determine the equivalent height to theoretical plate.

The entire schematic diagram of the CO_2 absorption process is illustrated in Figure 9. The flow sheet represents a continuous absorption/regeneration cycling process. Note that the reactions of these alkanolamines and CO_2 are mainly occurred by electrochemical reaction in the aqueous solution. Typical reaction mechanism of MEA and CO_2 are as in the following equations.

$$2RNH_2 + CO_2 + H_2O \leftrightarrow (RNH_3)2CO_3 \quad (1)$$

$$(RNH_3)2CO_3 + CO_2 + H_2O \leftrightarrow 2RNH_3HCO_3 \quad (2)$$

$$2RNH_2 + CO_2 \leftrightarrow RNHCOONH_3R \quad (3)$$

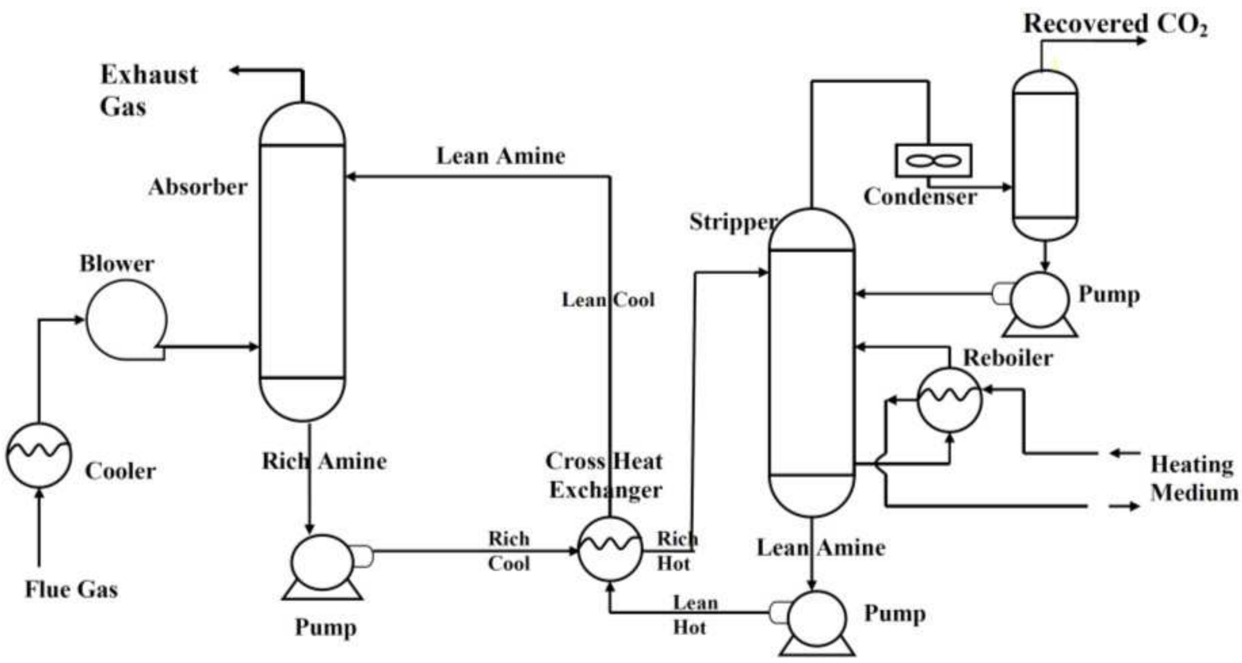

Fig. 9. Flowchart for CO_2 capture from flue gases using MEA, DEA and MDEA.

4. Results

Three different alkanolamines (MEA, DEA, and MDEA) were used in the simulation investigation of CO_2 capture in this work. Composition and thermal specifications of feed (flue gas and amine), entering the first tray at the bottom of the absorber are presented in Tables 1 and 2 respectively.

Component	Molar flow rate (kgmol/h)	Mol .fraction
N_2	7691.244	0.782
CO_2	1208.637	0.123
O_2	205.463	0.021
H_2O	728.561	0,074
Sum	**9833.906**	**1.000**

Table 1. Flue gas composition

Property	Flue gas	MEA	DEA	MDEA
Vapor fraction	1	00	00	00
Temperature *(°C)*	45.0	35.0	35.0	*35.0*
Pressure (kPa)	130.0	105.5	105.5	105.5
Mass flow (kg/h)	288351.8	596633.4	2010937.5	1990020.5

Table 2. Thermal specification of feed

Many other technologies used for CO_2 capture are based on the MEA process, with changes made in either solvent choice or absorption/stripping methodology. Specifications of the absorber for three types of amines are listed in Table 3. Type and amount of packing are selected so that the maximum recovery is obtained using the minimum consumption of amine. Composition of exit gas and rich amine leaving absorption column is presented in Table 4.

Parameter	MEA	DEA	MDEA
Section diameter (m)	5.944	6.096	6.401
Max flooding (%)	67.4 13	68.985	68.147
X-sectional area	27.745	29.186	32.178
Section height	29.871	42.999	61.086
Section ΔP(kPa)	2.727	10.114	12.072
ΔP per length (kPa)	0.112	0.288	0.242
Flood gas velocity (m^3/m^2h)	14798.038	9479.649	9141.451
Flood gas velocity (m/s)	4.111	2.633	2.539
HETP(m)	0.853	0.860	0.873
HETP correlation	Frank	Frank	Frank
Packing correlation	Robbins	Robbins	Robbins
Packing type	Gempak (metal structured) 0.75 A	Gcmpak (metal random) No._2	Gempak (metal structured) 2 A

Table 3. Specification of absorption column

Component	MEA		DEA		MDEA	
	Exit gas	Rich amine	Exit gas	Rich amine	Exit gas	Rich amine
N_2	99.995	0.005	99.982	0.018	99.98	0.02
CO_2	3.351	96.649	2.062	97.938	30.394	69.606
O_2	99.991	0.009	99.966	0.034	99.961	0.039
H_2O	6.62	93.38	0.459	99.541	0.434	99.566
MEA	0.226	99.774	Nil	Nil	Nil	Nil
DEA	Nil	Nil	Nil	100.00	Nil	Nil
MDEA	Nil	Nil	Nil	Nil	Nil	100.00

Table 4. Upstream and downstream composition of absorption column

As it can be seen almost all the CO_2 in flue gas is recovered by MEA and DEA (above 96 and 97% for MEA and DEA, respectively) through one stage, whereas MDEA amine is observed to be unsuitable for one stage CO_2 recovery (about 30% recovery). Rich amine at the bottom of the column is pumped to a heat exchanger then, achieving appropriate thermal specifications, it is introduced into the stripping column. Specifications of stripper for three types of amines are listed in Table 5.

Parameter	MEA	DEA	MDEA
Section diameter (m)	9.296	5.486	9.906
Max flooding (%)	69.887	50.882	69.777
X-sectional area	67.877	23 .641	77.070
Section height	20.497	18.288	14.922
Section ΔP (kPa)	4.865	16.835	3.242
ΔP per length (kPa)	0.290	----	0.266
Flood gas velocity (m^3/m^2h)	14736.444	----	12316.179
Flood gas velocity (m/s)	4.093	----	3.421
Estimation of pieces of packing	146080059.333	----	1345560.256
Estimation of mass of packing (kg)	292161.187	----	310513.905
HETP (m)	0.976	----	0.995
HETP correlation	Frank	Frank	Frank
Packing correlation	Robbins	Robbins	Robbins
Packing type	Levapacking (plastic) No._2	-------	Ballast Rings (metal). 3&1_2_inch

Table 5. Specification of stripping column

5. Discussion

A brief review on the associated problems of CO_2 emission, such as health and environment effects and the increasing trend of its emission, indicate the seriousness of the CO_2 capture in Iran's energy sector. The Iranian industry sector with about 26% of the total CO_2 emissions was the second major contributor in 2008, and the largest source was the petrochemical industry. The progressively increases of the emission along with its negative effects on the environmental impact, makes the capture of this greenhouse gas a very important issue. The observation of the fact that the combustion of fossil energy contributes with about 84 % to the CO_2 emission in Iran, the general acceptance of gas in contrast with coal or oil, and the advantages of developed technologies applied in the Iranian petrochemical industry, make it possible to take advantages of the Amine-based CO_2 capture in Iran. In order to capture CO_2 from flue gas in one of the petrochemical plants in Iran, three different alkanolamines were utilized in this work.

Today, most of the CO_2 used by the chemical industry is extracted from natural wells. As the extraction price is close to that for recovery from fermentation and other industrial processes, it may be that soon CO_2 recovered from electric energy generation could find a large application in the chemical industry.

To be able to compare the amine processes, the same general configuration of the process, feed composition and flow rate was applied for alkanolamine plant. The amount of CO_2 recovery, amine consumption, mechanical and operational characteristics of absorption column, CO_2 purity in stripper column effluent, required energy of stripper and mechanical and operational features of stripper were compared for three types of amines.

The amount of CO_2 recovery for three types of amines is represented in Figure 10. According to this Figure, CO_2 recovery for MEA and DEA are above 96 % while this value for MDEA is less than 70%. It means that MDEA is weaker than two other amines and cannot be used for one stage processes.

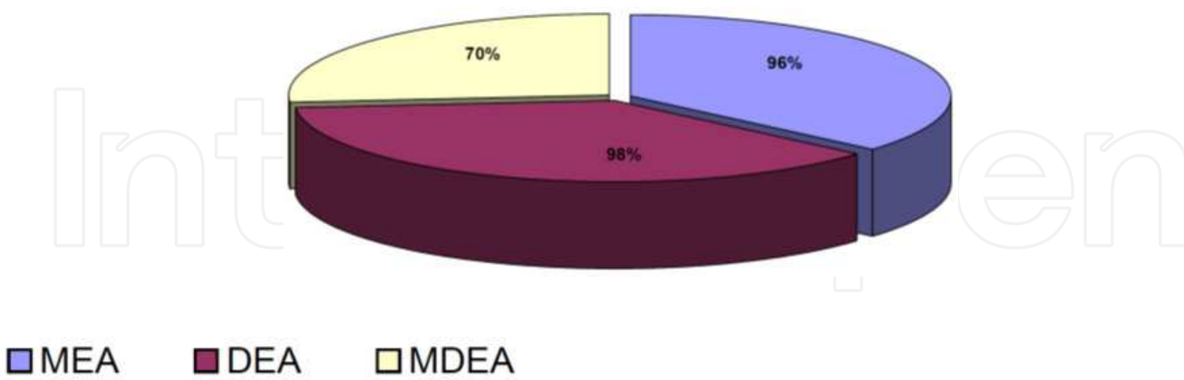

Fig. 10. CO_2 recovery (%) for three types of amine s process.

The amount of amine consumption for three types of amines is represented in Figure 11. As can be seen MEA process uses much fewer amines than other processes (about one fourth), i.e. this process is superior to other processes considering economic aspects. The low MEA consumption raises the reboiler duty substantially. The required pump power increases even more. Since the reboiler heat duty is the most important key to operating costs, this is a significant handicap (Chapel and Mariz, 1999).

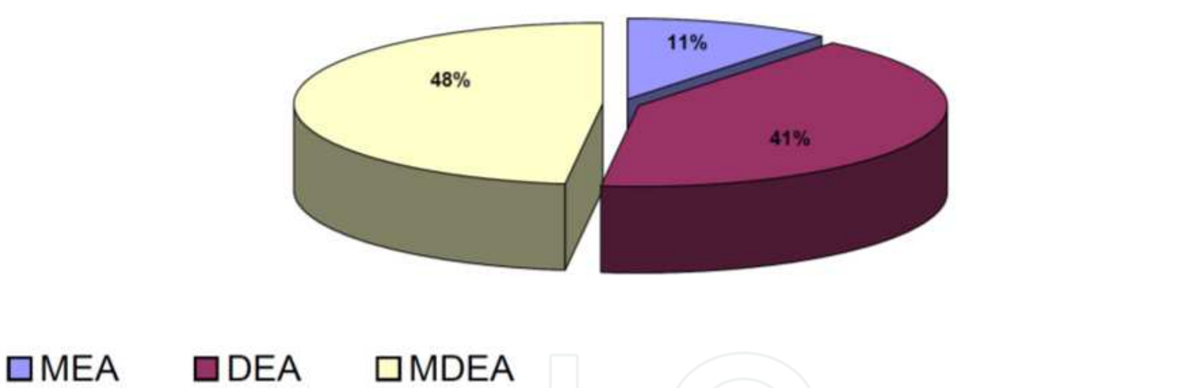

Fig. 11. The percentage of alkanolamine consumption (Kg/h) for three types of amines process.

Mechanical and operational characteristics of absorption column for three types of amines are almost the same, except to column height. Height of absorption column for MEA plant (30 m) is considerably lower than DEA (43 m) and MDEA (61 m) plants. Since, column diameter for all plants are the same, it can be concluded that MEA plant is better than others considering mechanical aspects.

Figure 12 indicates that, CO_2 purity in stripper column effluent is similar to all types of amines (above 97 %). Hence, this parameter could not be used as a criterion for selection of the optimum process.

As it is shown in the Figure 13, required energy of stripper for DEA plant is significantly smaller than other amine plants (about one tenth).

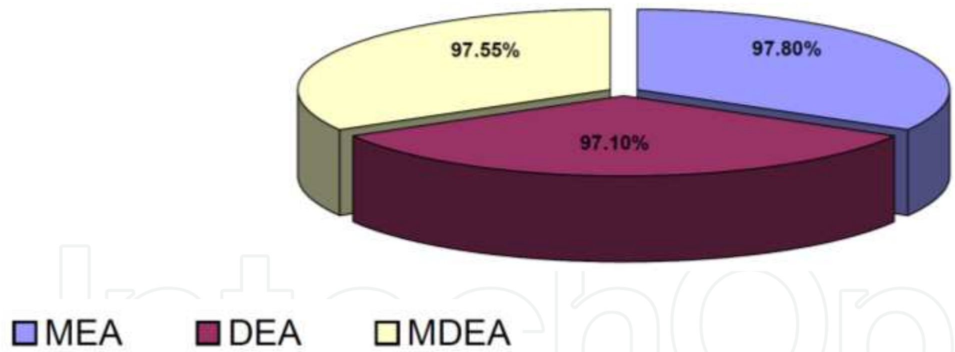

Fig. 12. CO_2 purity (%) for three types of amines process.

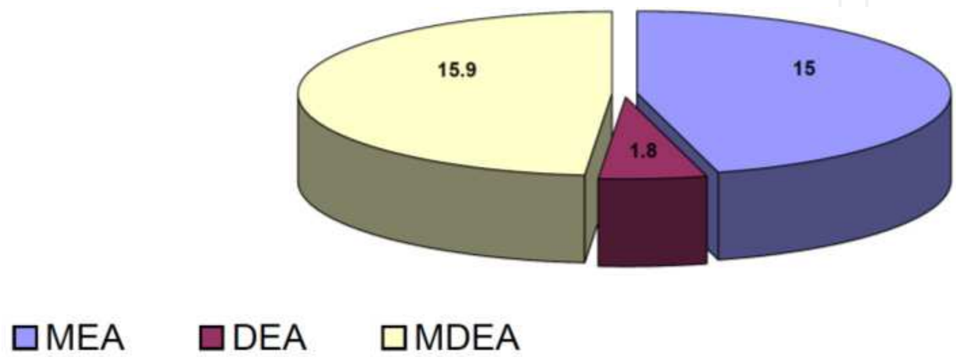

Fig. 13. Energy consumption (Kj/h) for three types of amines process.

Operational features of stripper are alike for all amine plants whereas mechanical characteristics are different to a certain extent. Diameter of MEA, DEA and MDEA stripper columns are 9.3, 5.5 and 10 m and heights of them are 20.5, 18.3 and 15 m, respectively. Taking all parameters into account, it can be concluded that the MEA plant is the best choice for separation of CO_2 from fuel gases.

The removal of CO_2 from flue gases using an amine depends on the gas-liquid mass transfer process. The chemical reactions that permit diffusion of CO_2 in the liquid film at the gas-liquid interface enhance the overall rate of mass transfer. Thus, the CO_2 removal efficiency in the absorber is a function of various parameters that affect the gas-liquid equilibrium (e.g., flow rates, temperature, pressure, flue gas composition, CO_2 concentration, alkanolamine concentration and absorber design). Similarly, the conditions and detailed design of the stripping column affect the energy requirements and overall performance of the system.

MEA is the most frequently used solvent for CO_2 absorption, and the greatest advantage of MEA is its relatively high loading. Two moles of MEA are needed for each mole of CO_2 absorbed, which represents the maximum equilibrium pickup and fixes the minimum circulation rate of MEA for completely treating a given quantity of acid gas.

6. Conclusions

In this chapter, the needs for CO_2 capturing were raised by presenting the negative health and environmental impacts of CO_2 emission in Iran. Direct relationship between fossil-fuel consumption and CO_2 emission was demonstrated in this study. Results show that CO_2 emission, especially from petrochemical plant, will have to be efficiently reduced. So, chemical absorption technology for CO_2 capture in a petrochemical plant has been selected in this study.

In the present work, CO_2 capture from fuel gases of one of the petrochemical companies in Iran using three alkanolamines (MEA, DEA and MDEA) was simulated and optimized. Specifications of absorber and stripper and composition of exit gas and rich amine leaving absorber were initially reported as simulation results. Then, these alkanolamines were compared considering some parameters such as CO_2 capture amine consumption, mechanical and operational characteristics of absorber and stripper, and CO_2 purity and energy consumption. It was found that, MEA and DEA are capable to recover almost all of CO_2 from flue gases. Amine consumption in an MEA plant is one-fourth of another amine plant where its energy consumption is the same as MDEA plant and ten times larger than DEA plant. Considering mechanical and operational characteristics, it was realized that MEA plant meets economic and aspects better than other amine plants. Finally, taking all parameters into consideration it was deduced that MEA is the best alkanolamine for separation of CO_2 from flue gases in this issue.

7. Acknowledgment

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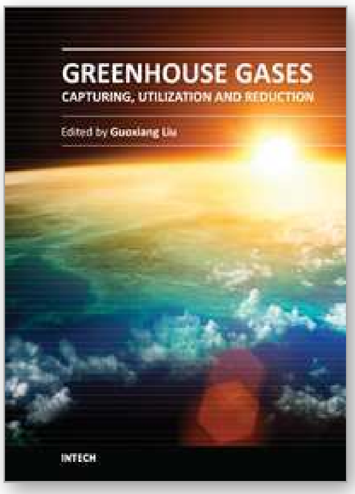

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Understanding greenhouse gas capture, utilization, reduction, and storage is essential for solving issues such as global warming and climate change that result from greenhouse gas. Taking advantage of the authors' experience in greenhouse gases, this book discusses an overview of recently developed techniques, methods, and strategies: - Novel techniques and methods on greenhouse gas capture by physical adsorption and separation, chemical structural reconstruction, and biological utilization. - Systemic discussions on greenhouse gas reduction by policy conduction, mitigation strategies, and alternative energy sources. - A comprehensive review of geological storage monitoring technologies.